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SPIN WAVES IN THE DILUTE HEISENBERG FERROMAGNET

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ABSTRACT

The energy of long wave length spin waves in dilute Heisenberg ferromagnets is calculated. It is shown that spin waves can exist only within an infinite cluster of magnetic spins and that such spin waves are mechanically stable. Heuristic arguments based on dimensionality of the infinite cluster imply that there is no thermal instability of the type discussed by Mermin and Wagner. Thus, the critical concentration for ferromagnetism is essentially equal to percolation concentration.

I. INTRODUCTION

Recently there has been increasing interest in determining the critical concentration of dilute Heisenberg and Ising ferromagnets.[1-7] The critical concentration x_c is the lowest concentration of magnetic atoms for which the system has long-range order. By analyzing the zero-temperature susceptibility as a series in powers of x, Elliott et al[4] concluded that x_c for Ising and Heisenberg systems should be the same for a given crystal structure and should be equal to the critical percolation concentration for that lattice.[5] Essentially this means that both systems have long-range order as soon as x becomes large enough to allow the formation of a single connected "infinite" cluster. Morgan and Rushbrooke[6] and Murray[7] have argued that while this result is understandable for Ising systems, it may not be correct for Heisenberg systems where there exist spin waves of arbitrarily low energy. In fact, for two-dimensional systems the Mermin-Wagner Theorem[8] shows conclusively that x_c cannot be the same for Ising and Heisenberg systems.

In this paper we study spin waves in the dilute Heisenberg system and discuss their relevance in determining x_c. A variational method is developed to calculate spin-wave energies. Using this method we conclude that there is no mechanical instability in the ground state. We also discuss the possibility of the kind of thermal instability that occurs in two dimensions. From qualitative arguments about the dimensionality of the infinite cluster we argue that x_c for the Heisenberg model is very close if not equal to x_c for the Ising system, in contrast to the results of previous authors.

II. THE UNRESTRICTED VARIATIONAL METHOD

We start with a general discussion of the model and the variational method. In the system we consider, Nx magnetic spins are arranged at random on a simple cubic lattice of N sites with Heisenberg interactions between spins on nearest-neighboring sites. Thus, for a particular configuration, θ, we write in the usual notation[3]

$$H(\theta) = -2J \sum_{n,m} \vec{S}_n \cdot \vec{S}_m; \qquad n,m \in \theta \tag{1}$$

where $n \in \theta$ means that for the configuration θ the lattice site n is occupied. Quantities of interest are evaluated by averaging over all configurations having a fixed concentration x of magnetic atoms.

For any configuration, the one spin-wave states are of the form,

$$|\psi_\alpha(\theta)\rangle = \sum_n b_\alpha(n,\theta) p_n(\theta)\, S_n^- |0\rangle, \tag{2}$$

where $p_n(\theta) = 1$, if $n \in \theta$ and vanishes otherwise. The coefficients $b_\alpha(n,\theta)$ are determined variationally from the energy. This procedure yields Nx eigenvalues $E_\alpha(\theta)$, which are averaged over configurations to give $\langle E_\alpha(\theta)\rangle$. After this averaging the system looks spatially uniform and α can be identified as a momentum: $\langle E_\alpha(\theta)\rangle \equiv E_k$. Evaluation of E_k using approximate wavefunctions yields an upper bound for the true E_k. Admissible trial wavefunctions must form an orthogonal set and, as we shall see, should not contain localized excitations.

Initially we ignore the latter restriction and develop the formalism given in Ref. 7. For sufficiently long-wavelength spin waves, arguments[9] based on the weakness of spin-wave scattering suggest the following first order choice for $b_\alpha(n,\theta)$ first given by Murray:[7]

$$b_k^{(1)}(n,\theta) = (2NSx)^{-1/2}\, e^{i\vec{k}\cdot\vec{r}_n}. \tag{3}$$

The resulting configuration-averaged energy, $E_k^{(1)} = 2JSa^2k^2x$, is characteristic of a molecular-field result and is expected to be good for $x \sim 1$. From a complete analysis of $b_\alpha(n,\theta)$ Murray obtained the following series in $(H_{kk'}/E_k^{(1)})$ for the energy correct to order k^2:

$$E_k = E_k^{(1)} - \sum_f{}' \frac{H_{kf}H_{fk}}{E_f^{(1)}} + \sum_{f\neq f'}{}' \frac{H_{kf}H_{ff'}H_{f'k}}{E_f^{(1)}E_{f'}^{(1)}} + \ldots, \tag{4}$$

where $H_{kf} = \langle k|H|f\rangle - E_k^{(1)}\delta_{kf}$. Retaining one and two terms in (4) yields curves a and b of Fig. 1, respectively.[7] Retention of the third term, which we evaluate as,

$$2JSa^2k^2\, \frac{(1-x)^2(1-2x)}{x} \left[\frac{0.09}{1-x} + \frac{0.77}{1-2x} + \frac{0.68}{x} + \frac{1-2x}{9x^2} \right] \tag{5}$$

yields curve c of Fig. 1. Generally, on truncating this series the variational significance of the result is lost. However a variational calculation of the energy to third order in $H_{kk'}/E_k^{(1)}$ yields the terms written in Eq. (4). From curve b it has been concluded[7,10] that $E_k(x)$ becomes zero at some x_o and that at this concentration the ferromagnetic ground state becomes unstable with respect to formation of long wavelength spin waves. Furthermore, x_o was identified as a lower bound for x_c, in spite of the fact that only curve c has variational significance.

Such an instability seems implausible to us. In fact, inclusion of the third order term changes the character of the curve to one which never becomes zero. This result agrees with the following simple intuitive reasoning. The ferromagnetic ground state is clearly mechanically stable. There is no physical reason to expect a condensation of spin-waves at zero temperature. In fact, the stiffness of an array of spins with respect to formation of spin-waves, measured by $D=\partial^2E(k)/\partial k^2$ is smallest for a linear chain oriented at random to k, for which $D=4JSa^2/3$. In the next section we show that if the variational principle is properly applied, results in accord with the above arguments are obtained.

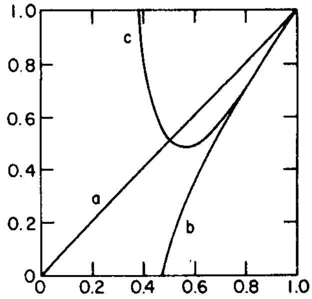

Fig. 1. $E_k(x)/E_k(1)$ vs x calculated using Eq. (4).

III. EXCLUSION OF LOCALIZED EXCITATIONS

At the outset we should distinguish between two regimes of concentration: one for which $x>x_p$, the other for which $x<x_p$. Spin-wave excitations can exist only above x_p and moreover, their wavefunction is confined to those spins in the infinite cluster. In the variational formulation described above the wavefunctions incorrectly included spin deviations in isolated clusters. To remedy this defect we adopt the revised first order wavefunction,

$$|\hat{k},\theta\rangle = (2NS\hat{x})^{-\frac{1}{2}} \sum_n e^{ik\cdot r_n} \hat{p}_n(\theta)S_n^-|0\rangle \tag{6}$$

in place of Eqs. (2) and (3). Here $\hat{x}$ is the concentration of spins in the infinite cluster and $\hat{p}_n(\theta)$ is unity if the nth site is occupied and is in the infinite cluster and is zero otherwise. The approximate spin-wave energy obtained from the wavefunction of Eq. (6) is

$$E_k^{(1)} = (\hat{k}|H|\hat{k})/(\hat{k}|\hat{k}) = 2JSa^2k^2x_1, \tag{7}$$

and forms an upper bound to the true $E_k(x)$. Here x_1 is the probability that the nearest neighbor of a spin belonging to the infinite cluster is occupied. The appearance of x_1 is understandable since zx_1 is the average number of occupied neighbors a spin sees in the infinite cluster and is clearly greater than zx. The result, Eq. (7) is shown as curve a in Figure 2.

This approximation also implies that the strength of the spin-wave pole in the transverse susceptibility, $\chi^{+-}(k,\omega)$, must vanish as the critical percolation concentration is approached. This is a direct consequence of restricting the spin-wave excitation in Eq. (6) to the infinite cluster. Formally this result can be obtained from the sum rule,

$$2\langle S_z\rangle_{H\to 0^+} = \int \mathrm{Im}\chi^{+-}(k,\omega)\,\frac{d\omega}{2\pi}\,. \tag{8}$$

As $H\to 0^+$, the magnetization of the finite-sized isolated clusters goes smoothly to zero, so that $\langle S_z \rangle_{H\to 0^+} = NS\hat{x}$ at zero temperature.

This formalism can be extended to higher order calculations. This is difficult because the occupation probabilities p_n are no longer site-independent. In Figure 2 the results of two improved variational calculations are presented. Curve b is an approximate evaluation of Eq. (4) in accord with the above restriction. Curve c results from inclusion of an additional variational parameter to improve the wavefunction near x_p. No claim regarding the accuracy of these results near x_p is made.

IV. LONG-RANGE ORDER IN DILUTE SYSTEMS

From the analysis of the previous sections it is clear that spin waves of finite stiffness may exist at all concentrations above x_p. However, this does not imply that the critical concentration x_c of the Heisenberg ferromagnet coincides with x_p. To illustrate this, consider the two-dimensional pure Heisenberg system. Qualitatively speaking, the two-dimensional system cannot support long-range order, because the density of states of long wavelength spin waves is too large. As a result, even at infinitesimal temperatures too many thermal spin waves are present and the ground-state order is destroyed. These considerations lead us to study the dimensionality of the infinite cluster near the critical percolation concentration. For example, if the infinite cluster for x near x_p is one-(or two) dimensional, long range order will not exist.

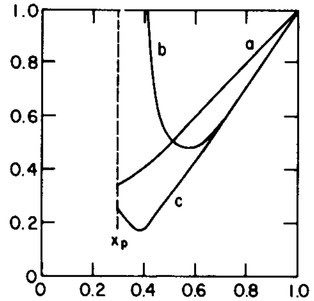

Fig. 2. $E_k(x)/E_k(1)$ vs x calculated using restricted variational wavefunctions.

In this section we give some heuristic arguments which show that "on the average" the infinite cluster is three-dimensional. It might appear that just above x_p the infinite cluster would be chain-like, since such a structure requires the minimum number of magnetic spins. However, we shall argue that the probability of its occurrence is vanishingly small. In other words, the entropy associated with a long chain is much less than that of a higher dimensional structure.

To see this we invoke an argument given by Kikuchi.[11] We divide the whole system into cells of equal size and shape. The size is large enough so that atypical configurations of cells occur with vanishingly small probability. In that case, all cells are more or less "typical" and hence interchangeable. Kikuchi has already argued that if infinite clusters exist, then in fact there is only one such. In each cell there are magnetic spins belonging either to the infinite cluster or to a finite cluster. From requirements of homogeneity and isotropy, part of the infinite cluster should belong to most cells. Even so, it is conceivable that there exists a chain winding its way through almost every cell. For this to happen, almost all cells would have to contain a segment of the chain which was not connected to segments in adjacent cells, as otherwise the structure would in fact be three-dimensional. However, the probability that these connections do not occur for any given cell is less than one. That they should rarely occur is therefore very unlikely.

Still another argument is based on the idea that if x is large enough for one infinite chain to occur, then an infinite number of such chains will occur. This is because chains occupy a volume of measure zero. Thus we might expect there to be many chains going in various directions, and arguing as above, we would expect that in reality the various chains are joined together.

A similar argument can be given which rules out the possibility of two-dimensional infinite clusters. If one such structure exists, then many will occur. In this case it is obvious that all such planes must intersect. Since one expects the orientations of the planes to be isotropically distributed over all solid angles, we again conclude that two-dimensional infinite clusters are connected to form a three-dimensional cluster.

Although these arguments are far from rigorous, they do indicate that the infinite cluster is three-dimensional. Accordingly we believe that the critical concentration for the Heisenberg ferromagnet must be close to, if not equal to, that of the Ising model.

This conclusion is in apparent contradiction to the analysis of high temperature expansions of the susceptibility.[6] However, such calculations may not reflect properly the dimensionality of the infinite cluster. It is true that the asymptotic behavior of the number of high temperature graphs as the graph size becomes infinite does define the dimensionality of the system. However, at present the practical limits on the computations are such that this asymptotic regime is completely inaccessible. Small graphs can only elucidate local properties and indeed locally clusters probably do look one-dimensional for x near x_p. In contrast, the critical concentration depends on dimensionality in a global sense.

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